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Influence of different deposition parameters on the performance of a Ti-Ta-C interface layer in SiC-fiber reinforced copper matrix composites

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Abstract. The mechanical properties of a SiC-fiber/copper matrix composite, reinforced with SCS-0 SiC-fibers (∅ 140µm, Specialty Materials), can significantly be increased by applying a Ti-Ta-C multilayer between fiber and matrix. This interlayer is deposited with a magnetron sputter device directly on the single fibers. By changing the deposition parameters of this sputter process the Ti-Ta-C interlayer can be optimized regarding fiber strength and fiber/matrix adhesion. Experiments with different deposition pressures, bias voltages and layer thickness' were performed to increase the bond strength and the ultimate tensile strength when compared to the Ti-Ta-C reference sample.

Introduction

Due to their attractive properties like high thermal conductivity and high temperature strength SiC-fiber/copper matrix composites are investigated as potential heat sink materials for future fusion experiments [1, 2]. The aim for this novel SiC-fiber/copper matrix composite is a thermal conductivity beyond 200 W m^{-1} K^{-1} with an acceptable ultimate tensile strength at temperatures around 820 K. One major challenge with this composite material are the different thermal expansion coefficients of the fibers and the matrix. Therefore an optimized fiber/matrix interface is needed. For the experiments two types of silicon carbide fibers were used: SCS-6 and SCS-0 fibers (Specialty Materials).

Both fiber-types are CVD-grown (chemical vapor deposition) with a diameter of about 140 µm. The difference between the two types is the fiber surface: the surface of the SCS-0 fibers is a stoichiometric SiC surface, the SCS-6 fibers have a 3 µm thin carbon-rich surface layer [3]. The interface optimization was done by coating the single SiC-fibers with a magnetron sputter device. Within a former study the SCS-6 based metal-matrix composite was optimized with a thin titanium interlayer sputtered between the fiber and the copper matrix [4]. With this interlayer the adhesion of fiber and matrix was about ten times higher. The interfacial shear strength increased from 6 MPa to 70 MPa. This SiC-fiber/copper matrix composite is the reference material for the actual investigation.

Preliminary experiments with a SCS-0 based metal-matrix composite showed a good performance with respect to adhesion and fiber strength for a Ti-Ta-C multilayer sputtered on the SiC-fibers. Starting with this "standard" fiber coating different deposition parameters (*bias voltage*, *deposition pressure* and *layer thickness*) were varied and continuously optimized. The coated fibers and the composite samples were investigated by single-fiber tensile tests, atomic-force microscopy (AFM), push-out tests and three-point bending tests.

Experimental

The different Ti-Ta-C coatings were applied to SiC fibers (SCS-0, ∅ 140 µm, Specialty Materials) using a magnetron sputter device (Discovery 18, Denton Vacuum) with two cathodes which can be operated in DC or RF mode. Before interlayer deposition, the fibers were ion-etched to eliminate surface impurities.
The Ti-Ta-C coating is a multilayer of different elements. Figure 1 gives a schematic overview of the layer system. On the surface of the fiber a only 40 nm thin tantalum layer was deposited followed by a 220 nm thin mixed Ti, Ta and C layer. The ratio between Ti and Ta within this Ti-Ta-C layer was 1/5. For a better bonding between this layer and copper two thin TiC and Ti-layers were sputtered (Fig. 1).

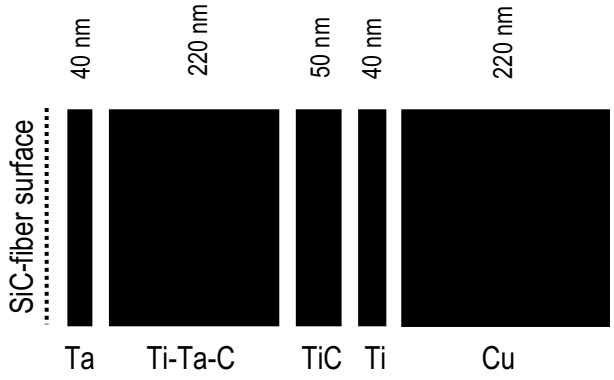

Fig 1.: Schematic view of the Ti-Ta-C multilayer system

To optimize the Ti-Ta-C multilayer the chamber pressure, the bias voltage and the layer thickness were varied. The parameters *chamber pressure* and *bias voltage* were applied to the whole layer system whereas the *layer thickness* was changed only for the mixed Ti-Ta-C layer. Table 1 gives an overview of the used combination of parameters.

	Chamber pressure	Bias voltage	Thickness (Ti-Ta-C)	Ti/Ta
Standard sample	3.8 x 10^{-6} mbar	0 V	220 nm	1/2.5
Sample 1	3.8 x 10^{-6} mbar	60 V	220 nm	1/5
Sample 2	3.8 x 10^{-6} mbar	110 V	220 nm	1/8
Sample 3	5.2 x 10^{-6} mbar	60 V	220 nm	1/5
Sample 4	8.6 x 10^{-6} mbar	60 V	220 nm	1/5
Sample 5	3.8 x 10^{-6} mbar	60 V	1000 nm	1/5
Sample 6	3.8 x 10^{-6} mbar	0 V	220 nm	1/1
Sample 7	3.8 x 10^{-6} mbar	0 V	220 nm	1/6

Tab. 1: Overview of the different deposition parameter combinations

After the deposition process the fibers were heat treated at 620 K and 920 K to investigate the temperature influence on the single-fiber properties. New formed phases after these heat treatments were determined by X-ray diffraction method (XRD) on planer samples.
To exclude the influence of preferential sputtering when using a bias voltage additional samples (*sample 6* and *7*) with different Ti/Ta-ratios where produced. The Ti/Ta-ratio was characterized by Rutherford-backscattering method (RBS, Ion: $^{4}He^{2+}$, Energy: 4 MeV). For the investigation of the fiber surface a atomic-force microscope (AFM) was used. The adhesion between the fibers and the matrix were measured with the push-out method which is explained elsewhere [4, 5]. The mechanical performance of the optimized SCS-6 metal-matrix composite and the reference material (SCS-0 metal-matrix composite) was tested by three-point bending experiments.

Results and Discussion

The surface of the fibers after deposition was characterized by an atomic force microscope. This investigation showed a significant influence of the grain growth and morphology by the different deposition parameters. Figure 2 shows a comparison between the standard (a) and three modified samples (b-d). In case of a high bias voltage of 110 V (Fig. 2b) the crystal seed density is strongly increased and the grains stay smaller. Additionally a preferred orientation of the grains and a high roughness can be seen. With a medium bias voltage of 60 V (Fig. 2c) the surface is very smooth with a low roughness and no visible grain orientation. In contrast to the 1 µm thick Ti-Ta-C coating – the surface showed clearly separated grains with deep grooves between the single grains (Fig. 2d).

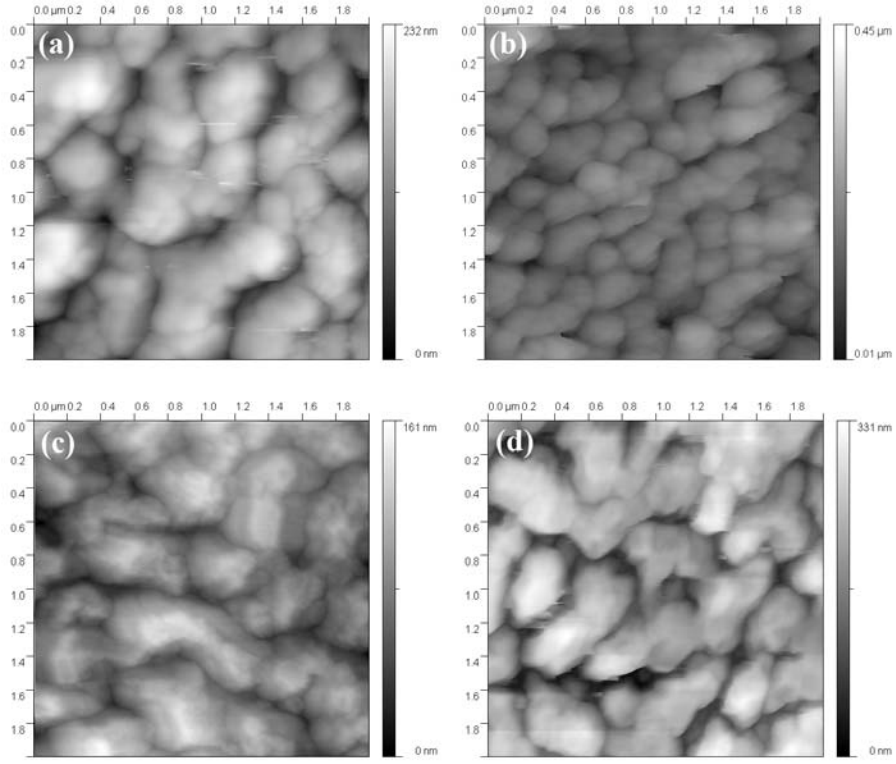

Fig.2: Different surface pictures made by atomic force microscopy (a) *standard sample*; (b) *sample 2*; (c) *sample 3*; (d) *sample 5*

The single-fiber tensile strength is strongly influenced by the different Ti-Ta-C coatings. Thereby the fiber surface hence the deposition parameters plays the major role. The highest ultimate tensile strength values (2500 MPa) after the heat treatment at 920 K for 1 h were measured for *sample 3* which showed the smoothest surface. The samples with sharply separated grains and deep grooves which means a great amount of potential surface defects (*sample 2* and *sample 5*) show lower tensile properties of around 2000 MPa after heat treatment at 920 K for 1 h.

To exclude the influence of preferential sputtering when using a bias voltage additional samples with different Ti/Ta-ratios where produced. Without any heat treatment the results of the single-fiber tensile tests showed higher ultimate tensile strength values for the *samples 6* (3100 MPa) and *7* (2800 MPa) when compared to the reference sample (2400 MPa). After the heat treatment at 920 K and a dwell time of 1 h the ultimate tensile strength of all three samples is around 1900 MPa. Therewith the tensile strength is not dependent on different Ti/Ta-ratios but on the created surface defects of the coated fibers.

The ultimate tensile strength for the optimized fiber coating (*sample 3*) after a heat treatment at 820 K and a dwell time of 480 h (3000 MPa) is about 800 MPa higher when compared to uncoated SCS-0 fibers (2100 MPa). Due to diffusion processes and a therefore lower intrinsic stress distribution within the coating the tensile strength of *sample 3* is higher after this long-term heat

treatment at 820 K. The tailored Ti-Ta-C coating increases not only the adhesion between fiber and matrix but also strengthens the fiber itself by reducing the surface defects. Table 2 gives an overview about the single-fiber tensile test results.

	ultimate tensile strength			
	290 K	**620 K, 1 h**	**920 K, 1 h**	**820 K, 480 h**
	[MPa]	**[MPa]**	**[MPa]**	**[MPa]**
Standard sample	2400	2200	1900	-
Sample 1	3050	3050	2300	-
Sample 2	2550	2400	2150	-
Sample 3	2750	**3200**	**2500**	**3000**
Sample 4	**3400**	3100	2150	-
Sample 5	2400	2000	1800	-
Sample 6	3100	2500	1900	-
Sample 7	2800	2700	1900	-
SCS-0 fiber	2100	2100	2100	2100

Tab. 2: Overview of the ultimate tensile strength of the different fiber coatings; maximum values for each temperature marked bold

The adhesion of fiber and copper matrix is not dependent on the deposition parameters or the fiber-surface. Due to chemical reactions this composite property is mainly influences by the "first" fiber coating. This can be seen by comparison of different "first" elements like Cr, Ti, W and tantalum based coatings directly deposited on the fibers. A chromium coating for example forms chromium carbides and diffuses strongly into the fiber [5]. This chemical reaction is the reason for a very strong adhesion. The tantalum based coatings (pure Ta and Ti-Ta-C) also chemically react with the fiber surface and form carbides and silicides. X-ray diffraction measurements showed a formation of Ta_2C and $TaSi_2$ at the interface of SiC and Ta.

In contrast the tungsten layer did not react with the fiber surface which was shown by X-ray diffraction method. As a result the adhesion between the tungsten coated fiber and matrix was at the same level like the uncoated SCS-0 fiber [6]. The push-out forces for the tantalum based coatings were four times higher when compared to uncoated SCS-0 fibers [6]. No differences were measured for the different tantalum-based coatings therefore a change in the deposition parameters did not significantly influence the adhesion of fiber and matrix [6].

To compare the performance of SCS-6 fibers with a titanium interlayer and the SCS-0 fibers with the optimized Ti-Ta-C multilayer three-point bending tests on the two different samples were performed. Figure 3(a-d) shows the fracture surfaces of the SCS-0 and the SCS-6 based metal-matrix composite after the three-point bending test. The different adhesion between the two fiber-types and the copper matrix is obvious. The SCS-0 fiber coated with the optimized Ti-Ta-C layer (*sample 3*) is even after the bending experiment bonded to the matrix whereas the titanium coated SCS-6 fibers lost the contact to the matrix. The SCS-6 fibers itself fail at the SiC/C interface. Figure (d) shows parts of the 3 µm thin carbon layer still bonded to the matrix. Due to the weak adhesion the SCS-6 based metal-matrix composite shows a more ductile fracture behavior.

Summary

The experiments show very clearly a significant influence of the deposition parameters on the properties of the Ti-Ta-C interlayer hence on the coated fiber properties. This means that not only the "first" element affects the ultimate tensile strength of the SiC-fiber but also the parameter setup. The most important parameter herein is the applied bias voltage during the deposition. This bias voltage influences the morphology of the growing coating which is, as shown, directly connected to the ultimate tensile strength. In contrast the impact of the different deposition parameters on the

adhesion is not measurable. The adhesion of fiber and matrix is mostly dominated by the “first” element which is deposited to the fiber and the potential chemical reactions.

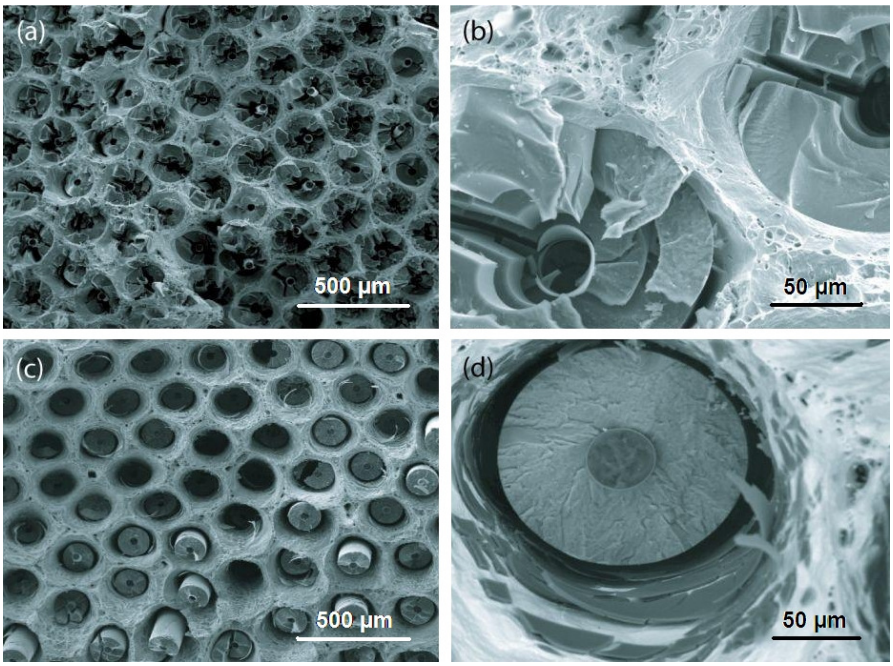

Fig.3: Comparison of the fracture surfaces of (a, b) the SCS-0 copper-matrix composite and the (c, d) SCS-6 copper-matrix composite

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References

[1] J.H. You and H. Bolt. Structural analysis of a plasma facing component reinforced with fibrous metal matrix composite laminate. Journal of Nuclear Materials, 329-333, 702 (2004).

[2] J.-H. You. Design feasibility study of a divertor component reinforced with fibrous metal matrix composite laminate. Journal of Nuclear Materials, 336, 97 (2005).

[3] S. Q. Guo, Y. Kagawa, Y. Tanaka, and C. Masuda. Microstructure and role of the outermost coating for tensile strength of SiC fiber. Acta materialia, 46, 4941 (1998).

[4] A. Brendel, C. Popescu, H. Schurmann, and H. Bolt. Interface modification of SiC-fibre/copper matrix composites by applying a titanium interlayer. Surface & Coatings Technology, 200, 161 (2005).

[5] T. Köck, A. Brendel, and H. Bolt. Interface reactions between silicon carbide and interlayers in silicon carbide-copper metal-matrix composites. Journal of Nuclear Materials, 362, 197, (2007).

[6] T. Köck, PhD-thesis, IPP-Report 17/6 (2008)